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**RELIABILITY OF METAL/GLASS-CERAMIC JUNCTIONS MADE BY SOLID STATE BONDING**

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**Résumé** - L'assemblage par diffusion dit par thermo-compression permet de réaliser des liaisons céramique-métal étanches à l'hélium. Ce procédé engendre cependant des contraintes résiduelles d'origine thermique sous l'action desquelles les défauts de la céramique peuvent se propager et conduire à la rupture différée de l'assemblage. Ce phénomène est mis en évidence sur des assemblages vitrocéramique/Al/Invar où une meilleure fiabilité est assurée par une réduction des défauts de surface.

**Abstract** - The solid state diffusion bonding leads to helium-tight ceramic-metal junctions. However this technique induces residual stresses due to expansion mismatches which may cause ceramic flaws to propagate hence junction delayed failure. This phenomenon is evidenced on glass-ceramic/Al/Invar junctions from which a better reliability is ensured by a reduction of surface flaws.

**I - INTRODUCTION**

The fabrication of electro-optical components of high quality and reliability lead us to achieve metal-ceramic junctions by solid state diffusion bonding. The dimensional stability required for such components dictated the selection of near-to-zero thermal expansion coefficients for the materials to be joined namely a glass-ceramic and a ferro-nickel alloy. The junction is expected to be helium-tight and to remain so after 20 000 hours' operation in addition it should hold temperature up to 200°C and resists thermal shock from this point down to -50°C. In order not to affect the glass-ceramic properties the joining process temperature should not exceed the transition point shown to be around 820°C, therefore techniques such as brazing or glass soldering are to be avoided.

Solid state diffusion bonding using aluminium as intermediate material has been proved in the past to be very efficient in the joining of glass, sapphire or alumina to metal /1/, /2/. This technique was lately extended to non-oxide ceramics and shown to allow heat and thermal shock resistant junctions /3/. However the high thermal expansion of aluminium and of metals relatively to ceramics is expected to yield residual stresses that may cause catastrophic failure after cooling or after thermal cycling.

The present study was undertaken after the observation of delayed failures of glass-ceramic/metal junctions without application of additional stress. It aims at the determination of the stress field thermally induced after the joining process and then allowing for the sensitivity to slow crack growth of the glass-ceramic the surface defects were suspected to play a major role on the junction reliability.

## II - EXPERIMENTAL

1. Joining technique. The solid state diffusion bonding consists in interposing a metal gasket between the ceramic and metal surfaces to be bound. The said gasket is then plastically deformed by rapidly squeezing the assembly at a temperature ranging from 0.5 to 0.9  $T_m$  ( $T_m$  = melting point of the intermediate metal). The bonding is due to various interface reactions depending upon the materials and the joining parameters.

In our case a low expansion glass-ceramic <sup>(1)</sup> ( $\alpha \approx 10^{-8} . ^\circ\text{C}^{-1}$ ) was sealed to invar-type ferro nickel alloy using an aluminium ring of hexagonal cross section. The surfaces to be in contact were ground flat up to a polish acceptable for helium-tightness and cleaned. Prior to joining the components were thermally treated in order to relax the residual stresses and to get some primary recrystallization.

The bonding achieved at about 500°C in flowing nitrogen added with 10 % hydrogen, under a compressive load ranging from 20 to 60 MPa. The strain rate ( $\approx 10^{-4} \text{ m.s}^{-1}$ ) is high enough to break the surface alumina on the ring. Bonding then occurs between fresh aluminium and the glass-ceramic via oxido-reduction and friction adhesion processes on one side and between aluminium and metal via diffusion on the other side. There might be some diffusion between aluminium and the glass-ceramic but it is hardly detectable by conventional microprobe analysis. In comparable experiments conducted elsewhere /4/ some diffusion was outlined on a very short range via SIMS analysis. This interface reaction aspect is under further investigation and will be discussed elsewhere.

The schematic of the technique is shown on Fig. 1, the details of the process have been presented elsewhere /5/, the aspect ratio of the aluminium ring was varied from 1 to  $\approx 20$  by the deformation process- and an electro-optical assembly thus made is also shown /6/.

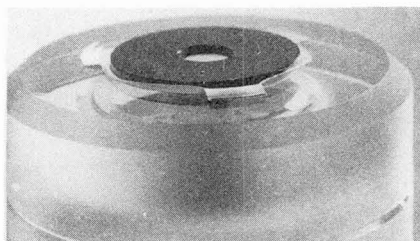
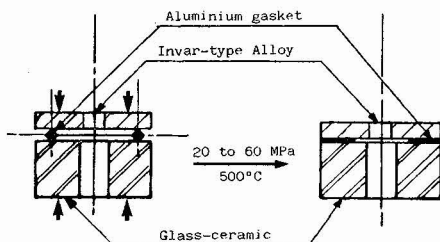


Fig. 1 - Schematic of the bonding technique and view of an electro-optical assembly.

2. Reliability of the junction. It has been observed that from a population of 40 junctions achieved according to the same bonding procedure 7 of those perished by delayed fracture of the glass-ceramic after different periods of time - from 1 to 600 hours - though all 40 were found to be helium-tight after joining. The remarkable fact is that all those 7 were obtained from glass-ceramic parts the surfaces of which had been only ground whereas all polished surfaces yielded junctions with lifetimes over 10 000 hours. Table I summarizes those observations, all survivors could resist the 200°C thermal shock test.

(1) Zerodur from schott : 70 to 75 wt %  $\beta$  euclerite and glass for balance.

Surface preparation	Defect maximum size $\mu\text{m}$	Number of junctions	Lifetime h
Ground	30	1	< 1
		1	3
		1	168
		2	240
		1	264
		1	576
		13	> 2000
			< 10 <sup>4</sup>
Polished	7	20	> 10 <sup>4</sup>

Table I - Junctions lifetimes observed after bonding vs surface preparation

Starting from these considerations we came up with the idea that delayed fracture could pertain to some crack growth phenomenon - as previously observed for soda-lime-silica glasses in moisture-containing atmosphere /7/ connected with the presence of defects resulting from the surface finish of the glass-ceramic. The observation of a fractured specimen such as shown in Fig. 2 is consistent with this idea, it can be seen that the fracture initiated from the surface and propagated in the body deviating towards the center hole of the specimen by contouring the zone underlying the interface.

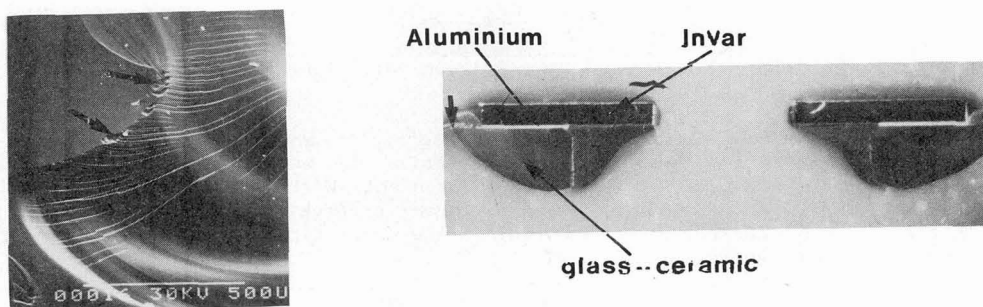


Fig. 2 - Fractography of the glass-ceramic and cross section of the fractured metal/glass-ceramic junction.

Since no additionnal stresses have been applied onto the specimens after joining one should suspect residual stresses resulting from the process to be responsible for the crack propagation in the glass-ceramic.

3. Mechanical characterization of the glass-ceramic. Since the above-mentioned stresses are expected to be complex, different testing techniques have been used to determine the failure stress of the glass-ceramic. Table II gives the values obtained vs the testing mode and comparable results allowing for the surface finish are also given. It is to be noted that the strength level is reduced by a factor of 2 when the surface is as ground instead of polished. Optical microscopy revealed defects of up to 30  $\mu\text{m}$  and 10  $\mu\text{m}$  in size respectively, which is consistent with the difference of strength level.

Testing mode	Surface preparation	Failure stress MPa	Weibull parameters $m$ $\sigma_0$	
Shear	Ground	65 $\pm$ 15		
	Polished	135 $\pm$ 26		
Compression	Polished	614 $\pm$ 14	7,8	43
3pt bending	Ground	157 $\pm$ 21		
biaxial 4pts bending	Ground	83 $\pm$ 11	6,7	80

Table II - Strength characteristics of the glass-ceramic vs surface preparation.

The critical stress intensity factor  $K_{IC}$  has been determined using three techniques, Vickers indentation - direct /8/ and post flexure strength /9/ - and double torsion /10/, values between 0,6 and 1,15 MPa. $\sqrt{m}$  were obtained.

The sensitivity to slow crack growth has been investigated using the double torsion technique /7/, /10/ with 80 x 40 x 5 mm<sup>3</sup> plates (Fig. 3) in atmospheres containing from 16 % to 100 % humidity. Direct visualization of crack extension was performed. Resulting  $V = A K_I^n$  curves that expresses the crack propagation rate  $\frac{da}{dt}$  vs stress

intensity factor  $K_I$  derived from the formula :

$$K_I = P W_m \left[ \frac{3(1+\nu)}{w d^3 d_n} \right]^{\frac{1}{2}}$$

where P is the applied load and  $\nu$  the Poisson ratio (see Fig. 3 for the other terms) are also presented.

A and n values were calculated by linear regression and found to be 0,71 and 16,8 respectively, these parameters thus characterize the sensitivity of the considered glass-ceramic in a moisture containing environment. Whether the latter is saturated or not does not affect to a great extent the crack propagation rate, this is consistent with the fact that the chemical potential at the crack tip is rather constant whatever the overall OH-concentration.

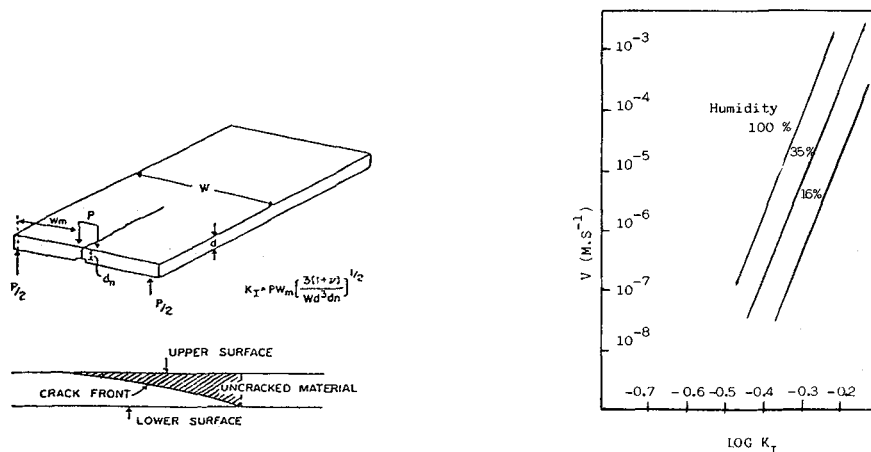


Fig 3 - Double torsion specimen used for the determination of  $K_I$  and  $K_I(V)$  curves obtained in various moisture - containing environments.

At this stage it is possible to predict the lifetime of the junction using the Weibull distribution parameters (see Table II), in the formalism derived from Griffith's Law.

Thus Time to Failure (t) vs Probability of rupture at constant applied stress ( $\sigma_a$ ) can be expressed as follow :

$$t = \frac{2 \sigma_o^{n-2}}{\sigma_a^n \cdot A(n-2) K_{IC}^{n-2}} \left[ \frac{\ln \frac{1}{1-p}}{\frac{S}{m+1}} \right]^m$$

where  $m$  and  $\sigma_o$  are the Weibull parameters,  $A$  and  $n$  the crack growth parameters and  $P$  the probability of rupture.

Taking  $m = 7,8$ ,  $\sigma_o = 43,4$ ,  $A = 0,71$ ,  $n = 16,8$ ,  $K_{IC} = 0,61 \text{ MPa} \cdot \sqrt{\text{m}}$ ,  $S = 240 \cdot 10^{-6} \text{ m}^2$

allow to plot Strength-Probability-Time (SPT) diagrams as shown on Fig. 4, which pertain to polished glass-ceramic specimens. They show for example that under a tensile stress of 40 MPa the glass-ceramic has 10 % failure probability after 1000 hours ( $\approx 4 \cdot 10^6 \text{ s}$ ).

The knowledge of the stress level and direction then appeared of prime importance to assess the reliability of the junction.

### III - STUDY OF THE RESIDUAL STRESS FIELD AFTER JOINING

The first approach consisted in considering the Evans' relationship applied for thermal elastic stresses in a body submitted to a thermal shock :

$$\sigma = \frac{E \alpha \Delta T}{1 - \nu}$$

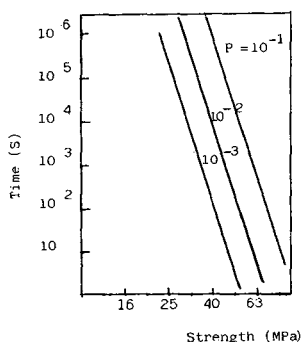


Fig. 4 - SPT diagrams for the glass-ceramic.

It can be shown that thermal stresses induced by the expansion mismatch after bonding in the joined assembly may be expressed as follow /5/.

$$\sigma = E \cdot \alpha \cdot \Delta T \cdot \frac{e}{h} \cdot K$$

where  $E$  = Young's modulus

$\alpha$  = mean expansion coef. of the metal

$e$  = thickness

and  $h$  = contact width

$\Delta T$  =  $T_{\text{process}} - T_{\text{reference}}$

$K$  = geometry factor bound to the metal substrate thickness.

This analytical formula allows the calculation of the maximum stress thermally induced at the interface. Depending upon the thickness of the metal substrate - from 0.5 mm to 1 mm - that affects the value of  $K$ , we have calculated stress values ranging from 50 to 160 MPa (the thicker the metal the higher the stress). Though this approach

gives some idea of the stress level it doesn't give any indication on both the distribution and the direction of stresses and doesn't take account of the behaviour of the intermediate aluminium layer - which seems more critical for in some cases the calculated stress is over the yield stress of the metal itself.

Finite element analysis was then carried out for further refinement of stresses evaluation using the values of  $E$ ,  $\nu$  and  $\alpha$  shown in Table III for the three components to be bound. The analysis was done for bidimensional and axisymmetrical modes, with linear displacement. The limit conditions were taken as follow :

- elastic behaviour for metal and ceramic substrates with no temperature dependence.
- allowance was made for the plastic behaviour of aluminium, considered as isotropic and non temperature-dependent.
- the stress is pure thermal with  $\Delta T = 500^\circ\text{C}$ .

The basic mesh corresponding to the actual electro-optical component is shown on Fig. 5 along with the resulting isostress lines. The stress analysis then shows that normal stresses are found near the outer cylindrical surface of the glass-ceramic substrate whereas compressive stresses are outlined around the central bore.

Materials	Young's modulus $E$ (GPa)	Poisson's ratio $\nu$	Thermal expansion coefficient $\alpha$ (20-500) $\text{K}^{-1}$
Invar	135	0,32	$6,5 \cdot 10^{-6}$
Al	78	0,3	$24 \cdot 10^{-6}$
Glass/ceramic	95	0,25	$\approx 10^{-8}$

Table III - Physical properties of the three materials to be solid state bonded.

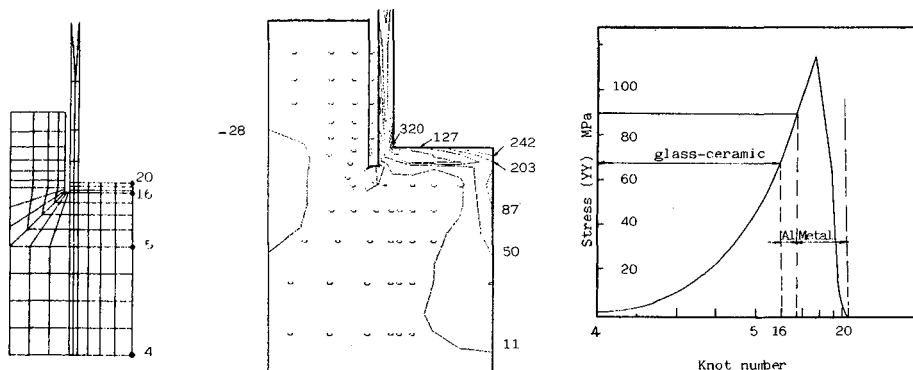


Fig. 5 (Left) - Stress field analysis of the electro-optical assembly after solid state bonding main stress in MPa, negative for compressive. (Right) Tensile main stress calculated along the external generatrix.

The analysis of principal stresses located at the interface revealed high levels of stress in the aluminium layer, which are consistent with the plasticity hypothesis on stress relaxation. As for the glass-ceramic substrate values ranging from 20 to 80 MPa were found depending upon the stress mode and the location. Therefore, although the stress level is reduced by the creep of aluminium, the glass-ceramic appears to be the siege of residual stresses likely to induce slow crack growth.

As a conclusion we could confirm that the observation of delayed fracture of glass-ceramic substrates after joining is consistent with the fact that under the simultaneous action of humidity in ambient air and residual stresses a slow crack propagation is enhanced. The influence of the surface preparation is of prime importance on the crack initiation and a careful polish may be sufficient to improve

lifetime to a large extent. The reliability of junctions achieved by solid state bonding can be better assessed through a finer stress field analysis.

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